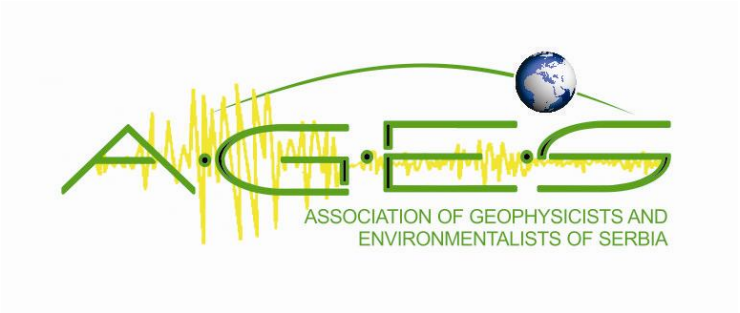

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Improvement of the research methodology of groundwater contamination by petroleum hydrocarbons in Serbia

Nenad Marić*, Stanko Sorajić, Jelena Zarić, Marko Vanić

Summary

During the last 30 years the Republic of Serbia has had several major accidents with petroleum hydrocarbons which have caused significant negative effects on groundwater and geologic media. Data collected at locations of mentioned accidents are used this paper in order to present the methodology which was applied in this type of research in Serbia. Beside mentioned methodology as it basis, particular tendency of this paper is to provide insight into the basics of bioremediation and natural attenuation as emerging approaches for mentioned issues. The application of these approaches in groundwater quality improvement in Serbia is at the beginning. This paper aims to emphasize the necessity of research methodology improvement in order that hydrogeology can provide full contribution to the application of bioremediation methods in field conditions.

Introduction

Groundwater represents about 98% of the available fresh water of the planet (Alvarez and Illman, 2006). Contribution of groundwater in the public water supply of the most European countries is dominant. Similarly to these countries, about 80% of the public water supply of the Serbia comes from groundwater (Polomcic et al., 2011). Considering the extensive use of petroleum hydrocarbons, this group of compounds is among the most common groundwater contaminants. According to Vujasinovic and Matic (1995) on the teritory of the Republic of Serbia several major accidents has happened which have caused contamination of groundwater and geologic media by petroleum derivates, see black points in Figure 1. As the basis of this paper, united results of research performed at mentioned locations are presented, in order to provide insight into applied methodology.

On the other hand, during the last 20 years research of the environmental impacts of petroleum hydrocarbons have been significantly improved. This improvement has caused better understanding of the behavior of these compounds in field conditions, as a necessary precondition for the selection of appropriate remediation methods. All this has resulted in the necessity to extend approach for mentioned environmental issues, from entirely hydrogeological used so far, to much more complex interdisciplinary approach. In this context basics of *in situ* bioremediation methods and their significance for the preservation of groundwater quality are given.

Year of accident	Location	Description
1979.	TE "Obrenovac"	leakage of the heavy oil – Sava river and coast line
1982.	Uz.Pozega	leakage of the diesel at the railway station
1983.	TE "Kosovo"	leakage of the phenol in Ibar river
1984.	Makis – Beograd	accident of cisterns with xylene
1985.	Dunav stanica	leakage of the diesel
1986.	Lucani (Cacak)	leakage of the heavy oil
1988.	Bozdarevac (Barajevo)	accident of tank wagons with diesel
1992.	Usce (Kraljevo)	leakage of the heavy oil at railway
1992.	Naumovicevo (Subotica)	leakage of the diesel
1993.	Vitanovac (Kraljevo)	leakage of the kerosene

Figure 1. *Locations of major accidents with petroleum derivates in Serbia (Vujasinovic and Matic, 1995)*

Data – Methodology

During the last 30 years the Republic of Serbia has had several major accidents which have caused contamination of groundwater and geologic media by petroleum hydrocarbons. Data collected at these sites are used in this paper in order to provide insight into methodology which was applied. The quantities of contaminants in mentioned accidents were significantly different, from about 45 tones in Makis, up to 540 tones in Vitanovac. The common thing for all these accidents is that they have significantly endangered groundwater and sediments in the area around the location of accident. This is a consequence of the construction of infrastructure facilities without considering of their impact on the environment and providing adequate conditions for its protection. The most obvious example for this is the construction of the freight station in the sanitary protection zone of Belgrade source in Makis.

Works performed in order to reduce negative effects of these accidents could be defined as investigation-remediation works. They were performed immediately after every accident in order to collect as much more of information and to enable as faster remediation. These works were concepted in a way to define structure of terrain, groundwater flow directions and spatial distribution of plume (piezometers, exploration wells, trenches), along with eventual prevention of contaminant migration in an undesirable direction. From the chemical aspect contaminant concentrations in groundwater and sediments were measured, with exceptions of the research at the locations of Usce and Bozdarevac where more detailed chemical analysis were performed. Applied remediation methods considered primarely of contaminant physical removal, whether from groundwater (pumping out) or taking of contaminated sediments to safe location. More detailed review of applied remediation methods is given by Vujasinovic and Matic (1991). By using this method significant quantities of contaminants were removed from contaminated aquifers, from about 17 tones in Makis to approximately 200 tones in Vitanovac. Due to their significance, results of these works have been published in international scientific journals. According to the achievements at that time, remediation works were unable affect the part of the contaminant which remained attached to the sediments in the subsurface, which has made some of these sites long time sources of contamination. Considering this fact and the latest world achievements, it is necessary to improve methodology of this type of research in Serbia through application of bioremediation methods.

Bioremediation and natural attenuation

Bioremediation is one of the remediation treatment processes which uses microorganisms that are naturally present at contaminated sites, in order to degrade harmful substances into less harmful or completely harmless (Kresic et al., 2006). From the aspect of preservation of groundwater quality, engineered bioremediation (biostimulation and/or bioaugmentation) and intrinsic bioremediation (natural attenuation) considered can be as significant. Success in the application of these methods in field conditions in a large extent depends on the contribution of hydrogeology. However, these methods are not universally applicable and may be insufficiently effective in inappropriate environmental conditions or when applied to "resistant" contaminants. The requirements for application of bioremediation methods are shown in a pyramidal structure in Figure 2 (Alvarez and Illman, 2006).

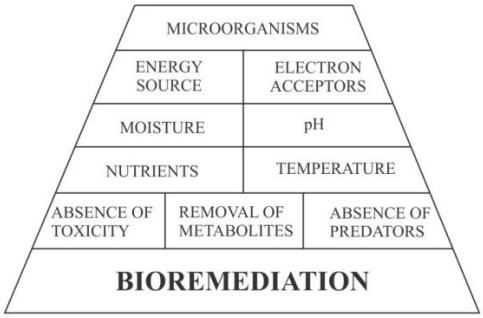

Figure 2. *Requirements for bioremediation methods (Alvarez and Illman, 2006)*

As can be seen, for the application of these methods there are several requirements. Beside the presence of microorganisms with capacity to synthesize enzymes which are able degrade harmful substances it is necessary that environmental conditions are favorable for their development. Considering the mentioned cases of contamination by petroleum hydrocarbons in Serbia, the role of natural attenuation is emphasized in this paper. According to U.S. EPA (1999) natural attenuation refers to the reliance on natural attenuation processes to achieve site-specific remediation objectives within a time frame that is reasonable compared to that offered by other more active methods. Same authors refer that natural attenuation processes include a variety of physical, chemical, or biological processes that, under favorable conditions, act without human intervention to reduce the mass, toxicity, mobility, volume, or concentration of contaminants in soil or groundwater. These in-situ processes include biodegradation, dispersion, dilution, sorption, volatilization, radioactive decay, chemical or biological stabilization, transformation, or destruction of contaminants. Biodegradation (degradation of organic compounds mediated by microorganisms) represents the dominant destructive mechanism by which amount of contaminants reduces.

Groundwater quality and bioremediation methods in Serbia

Application of bioremediation methods in preservation and improvement of groundwater quality in Serbia is at the begining. Aim of this paper is to point out the neccesity of improvement methodology of hydrogeological research related to the groundwater contamination by petroleum hydrocarbons. This improvement primarely implies that hydrogeological research should be concepted in a way to provide conditions for application of bioremediation methods. In this context measurements of contaminant concentrations should be followed with monitoring of degradation products and geochemical parameters. Due to the mentioned cases of historical contamination in Serbia, parameters relevant for the monitoring of the processes of natural attenuation are given in this paper. Complete suite of geochemical parameters for the monitoring of natural attenuation is given by Weidemeir (1995, 1999). As in the case with other specialized research in hydrogeology, measurements of these parameters in field conditions and their general interpretations can be carried out by hydrogeologists. Detailed overview of geochemical parameters and guidelines for their interpretation is given by Nielsen (2006), while a short overview is given in Table 1. An example of this improved research which included measurements of mentioned parameters was performed in 2011 at the location of Vitanovac.

Geochemical parameter	Description
Dissolved Oxygen	The favored electron acceptor for aerobic biodegradation, whose concentrations decrease during time due to its use by microbes. This results in creation of anaerobic aquifer conditions.
Nitrate	Electron acceptor for anaerobic biodegradation via denitrification. Active process will cause decrease of nitrate concentrations in groundwater.
Sulfate	Electron acceptor for anaerobic biodegradation via reduction of sulfate to sulfide. During this process concentrations of sulfate will be decreased.
Fe (II)	Significant factor for abiotic reactions, its increased contentrations indicate that anaerobic biodegradation has occured via Fe(III) reduction.
Methane	Product of biodegradation of organic carbon and indicates strongly reducing conditions and particularly process of reductive dechlorination.
Temperature	Generally affects solubility of oxygen and other geochemical parameters, but rarely significantly limiting factor for the processes of degradation.

pH	Indicator of the character of environment, microbes generally prefer neutral or slightly alkaline conditions.
Conductivity	Measure of the ability of a solution to conduct electricity, directly related to the concentration of ions in solution. Significant indicator of groundwater flow path.
Oxidation – Reduction Potential	Measure of the tendency of a solution to accept or transfer electrons. Lower values indicate reduction conditions. Can be helpful in spatial defining of contaminant plume.

Table 1. *Geochemical parameters for monitoring of natural attenuation (adapted from Nielsen (2006)*

Conclusion

Beside many years of experience in solving problems of groundwater contamination with petroleum hydrocarbons in Serbia it is necessary to improve methodology of hydrogeological research related to these issues. This improvement is necessary if we consider insufficient effectiveness of conventional remediation methods and emerging worldwide application of bioremediation methods. In this context, the contribution of hydrogeology in application of bioremediation methods must be crucial in the interpretation of environmental conditions and interaction between environment and contaminants. Considering the importance of bioremediation methods in field conditions for the preservation of groundwater quality, it is necessary to upgrade the existing knowledge of hydrogeological experts through the creation of multidisciplinary teams and collaboration with experts from other areas. Improvement of the methodology is necessary in order that hydrogeology could give full contribution to the application of bioremediation methods in field conditions.

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